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Absence of Magnetism in HCP Iron-Nickel at 11 K

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Density functional theoretical (DFT) calculations predict an antiferromagnetic ground state for the high-pressure hcp phase of Fe and for hcp Fe-Ni. We performed synchrotron Mossbauer spectroscopy (SMS) on an hcp-phase $Fe_{92}Ni_8$ alloy at 28 GPa and 11 K. The SMS spectrum showed no hyperfine magnetic field, in disagreement with the DFT calculations. Our results are consistent with significant errors in the GGA density functional, leading to an erroneous prediction of a large HMF. Alternately, these results indicate the presence of quantum fluctuations with periods shorter than the lifetime of the nuclear excited state, prohibiting the detection of magnetism by the SMS technique.

Elemental iron, which has the body-centered cubic (bcc) crystal structure at ambient temperature and pressure, transforms to the hexagonal-close packed (hcp) phase at a pressure of approximately 13 GPa. An antiferromagnetic (AFM) ground state has been predicted repeatedly for the hcp phase, but experiments have never detected the presence of a hyperfine magnetic field (HMF), related to the net spin density at the nucleus, implying the absence of static magnetic moments or magnetic order. Recent density functional theory (DFT) calculations have suggested that the vanishing HMF in hcp iron can be explained by cancellation of a large core electron polarization by an equally large itinerant electron polarization in the antiferromagnetic state. DFT calculations for the AFM structure have shown markedly better agreement with equation of state and elasticity measurements than nonmagnetic calculations. We tested the idea that ε-iron is antiferromagnetic, yet exhibits no hyperfine field owing to the cancellation of up and down spin densities at its nuclei. If indeed such a balance between core and conduction electron polarization exists, a magnetic perturbation should disrupt it, producing measurable hyperfine magnetic fields.

An alloy of nominal composition $Fe_{92}Ni_8$ was made by arc-melting. High-pressure energy-dispersive x-ray diffraction (EDXRD) was performed at NSLS beamline X17C to identify the pressure of the bcc to hcp phase transition, which occurred between 9 and 10 GPa. Full-potential DFT Linearized Augmented Plane Wave (LAPW) calculations were performed in the generalized gradient approximation (GGA) with the Wien2k software package. A 16-atom supercell was constructed, for the composition Fe_7Ni, shown in schematic in **Figure 1**. The antiferromagnetic structure was found to be more stable than the nonmagnetic structure, and HMF as high as 6.6 Tesla were found. Synchrotron Mossbauer spectrometry (SMS) experiments were performed on the sample of $Fe_{92}Ni_8$ at beamline 16 ID-D of HPCAT at the Advanced Photon Source at a pressure of 21 GPa and a temperature of 11 K. SMS detects hyperfine magnetic fields by the evidence of quantum beats, interference patterns of emitted gamma rays in the time domain. No quantum beats were detected in the spectrum emitted from the sample, in clear disagreement with the calculated results. Using the fitting routines in the program CONUSS, theoretical curves based on the calculated HMF were generated, and overlaid

Authors (from left) Michael Hu, Rebecca Stevens, and Alexander Papandrew prepare to lift a dewar of liquid Helium at HP-CAT.

with the data. These curves are shown in **Figure 2**.

There are two probable explanations possible for the disparity between the experimental and the calculated results. An overestimation by 20% of the strength of exchange coupling by the GGA functional results in a large change in the calculated magnetic moment, from 1.077 μ_B/atom to 0.053 μ_B/atom. Alternatively, quantum spin fluctuations thought to be responsible for superconductivity in ε-iron below 2K may be faster than the Mossbauer lifetime, inhibiting detection of a hyperfine field. The hcp lattice is geometrically frustrated with respect to antiferromagnetism, and it is known that fluctuations play an important role in the physics of many frustrated antiferromagnets. Spin fluctuation rates in the GHz range have been identified in these materials and cannot be discounted for the afmII or any other AFM spin structure for ε-Fe.

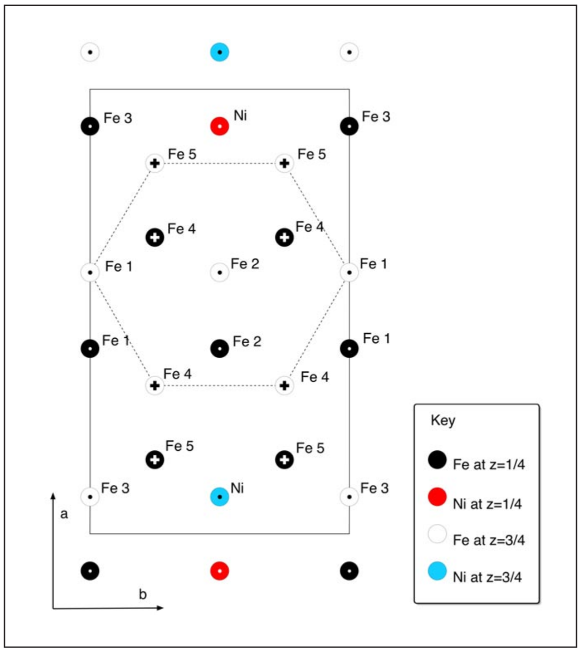

Figure 1. *The Fe_7Ni calculational supercell with the afmII spin structure. Crosses denote a spin orientation pointing into the page, while circles indicate spin pointing out of the page.*

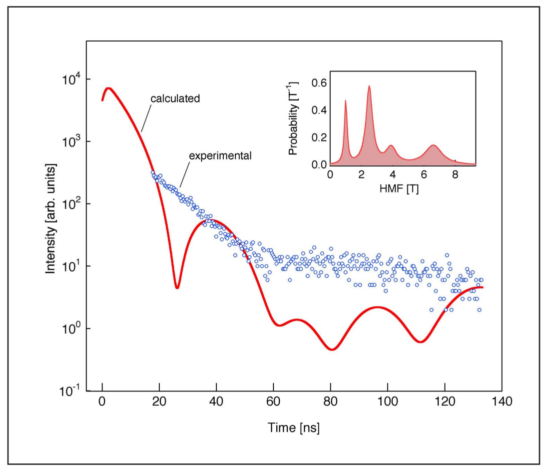

Figure 2. *Experimental data from hcp Fe92Ni8 at 11 K (blue) and a simulated SMS spectrum generated with CONUSS (red) based on DFT calculations of HMF in Fe_7Ni. Inset: The HMF distribution for the simulated spectrum.*